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IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re application	:	Satoshi Murouchi, et al.
Application No.:		10/519,046
Filed	:	August 19, 2005
Confirmation No.	:	6619
For	:	WHOLLY AROMATIC LIQUID CRYSTAL…
Examiner	:	Michael B. Nelson
Attorney's Docket	:	AK-481XX

TC Art Unit: 1794

* *

Declaration of Yoshiharu Iwasaki

Via EFS-Web
Commissioner for Patents
P.O. Box 1450
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Sir:

I, Yoshiharu Iwasaki, declare as follows:

1) I am currently Deputy General Manager of the Advanced Polymers Department of the Chemicals Division of Nippon Oil Corporation. My responsibilities have included the management of the LCP business of Nippon Oil Corporation since April of 2005. I also worked on production process development of polyethylene for 2 years, catalyst development for polypropylene for 3 years, and research and development for resin materials for automobiles for 7 years. My work has also included research and development for the injection molding of resin materials for 4 years and the

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development of LCP for 5 years. I am named as inventor on numerous granted patents, including 12 Japanese patents (e.g., JP 1,933,949, 2,050,588, and 3,506,539), 4 European patents (EP 267,794, 471,078, 466,930, and 465,664), and 4 U.S. patents (U.S. 5,266,362, 5,190,704, 5,200,122, and 5,240,973).

2) I am familiar with U.S. Patent Application Ser. No. 10/519,046, and I have read the application and examined the drawings for the application. I understand that this application claims priority to Japanese Patent Application No. 2002-185354 filed June 25, 2002.

3) It is known to a person of skill in the art of polymer resins that a mathematical relationship exists between the dielectric constant for a resin composition and the dielectric constants of the components of the composition. That mathematical relationship is the following:

$$\log \varepsilon = X_1 \log \varepsilon_1 + X_2 \log \varepsilon_2$$

In other words, the ordinary skilled artisan at the priority date of the 10/519,046 application would have expected that the

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logarithm of the dielectric constant of a product made by molding an organic resin composition is represented by the sum of the product of the volume fraction of each component and the logarithm of the dielectric constant for each component. This relationship is described on page 283 of "Theory of Solid Dielectrics" published in 1960 by Syoten OKA (Japanese version with English translation attached as Exhibit A). Specifically, ε represents the dielectric constant of the composition; ε_1, ε_2, etc., represent the dielectric constants of each of the components; and X_1, X_2, etc., represent the volume fractions of each of the components. This equation is thought to also apply for a resin composition comprising a liquid crystal polymer (hereinafter "LCP") and inorganic fillers, including inorganic spherical hollow material such as glass balloons (hereinafter "GB").

4) According to the above relation, the logarithm of the dielectric constant for a resin composition comprising LCP and inorganic spherical hollow material such as GB is the sum of the product of the volume fraction and the logarithm of the dielectric constant for each of the components (namely LCP, GB, and glass (fractured GB)). Therefore, it can be expected that the logarithm of the dielectric constant for the mixed resin composition (and

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also the dielectric constant itself) increases monotonically as the fracture rate of hollow material (i.e., GB) increases. Qualitatively, this can be understood as the steadily decreasing contribution of air, in the form of intact GB, as the GB fracture rate increases.

5) Thus, the ordinary skilled person at the priority date of the 10/519,046 application would have understood and expected that the dielectric constant for an LCP resin composition containing GB would monotonically increase as the fracture rate of GB increases.

6) Such an ordinary skilled person also would have expected that the dielectric constant for a molded product obtained by injection molding an LCP resin composition containing GB increases continuously over the range of fracture rate of 0.045 to 0.094, as recited in claim 1 of the 10/519,046 application.

7) I have reviewed Graph 1 that was submitted with the Applicant's response to USPTO on January 14, 2009 by the Applicant. Graph-1 shows a distribution of data in which no points are represented in the range of fracture rates from 0.05 to 0.08.

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However, the ordinary skilled person would have expected a continuous increase of dielectric constant over the range of fracture rates from 0.05 to 0.08, even though that range is not populated with data points. This is because, as explained in paragraphs 3) through 6) above, the ordinary skilled person would have expected the dielectric constant for an LCP resin composition containing GB to increase continuously as the fracture rate of GB increases, even if there had been no points between the endpoints of 0.045 and 0.094. From my professional point of view, it cannot be expected that the dielectric constant of a molded product obtained from an LCP resin composition comprising GB and having a fracture rate as recited in claim 1 of the 10/519,046 application would depart drastically from the relationship shown in Graph 1 and exceed a value of 3.0. There is no basis to expect such a result.

8) In conclusion, the ordinary skilled person at the priority date of the 10/519,046 application would have readily assumed that the dielectric constant of a molded product obtained by injection molding an LCP resin composition comprising the amount of inorganic spherical hollow material recited in claim 1 of the 10/519,046 application would be less than 3.0 over the

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entire range of fracture rate of the inorganic spherical hollow material of 0.045 to 0.094, even though this is not verified in the Examples over some portion the range. One of ordinary skill in the art would have appreciated that the inventors were in possession a molded product having a dielectric constant of 3.0 or less and a dielectric dissipation factor of 0.04 or less even at fracture rates in the middle section of the claimed range of fracture rates (e.g. 0.05-0.08).

I hereby declare that all statements made herein on personal knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Signed this 18 day of September, 2009.

Yoshiharu Iwasaki

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EXHIBIT A

Translation of "Theory of Solid Dielectrics" issued in 1960, Syoten Oka, Osamu Nakata, relevant part on page 283

§89 Dielectric constant of a mixture

Logarithmic relational expression is mostly approved for a mixture of rutile powder and other ceramics and/or organic resins and/or liquids.

$$\log \varepsilon = X_1 \log \varepsilon_1 + X_2 \log \varepsilon_2$$

wherein ε_1, ε_2: dielectric constant of each component and X_1, X_2: volume fraction thereof.

Generally it is very convenient if a dielectric constant of a mixed substance is calculated from components' dielectric constant or if other component's dielectric constant can be calculated knowing a dielectric constant of mixed substance and a dielectric constant of one component. Not limiting with a dielectric constant, these kinds of subjects for various kind of physical property values have been studied since long time among physicists.

に対して電子分極 p_e と原子分極 p_a を求め，表 11.2 の結果を得た．

ε_0 は電子分極による高周波誘電率（屈折率の 2 乗）で，p_a は $1-p_e$ から求める．表によればルチルは最大の電子分極で特徴づけられるが，原子分極は最大ではない．しかし全分極はやはりルチルが最大である．ゆえにルチルの高誘電率は電子分極（屈折率）の異常に大きいためと考えられる．なおルチル，板チタン石，鏡鉱石の誘電率の相違は密度の相違で大体説明される（表 11.3）．

表 11.3　TiO_2 の三態の密度，誘電率および分子分極

	ルチル	板チタン石	鏡鉱石
密　度 (g/cm³)	4.25	4.11	3.87
誘　電　率	114	78	31
分子分極 (cm³)	18.2	18.7	18.8

一般にイオン結晶の誘電率の温度係数は二つの部分から成る[13]．第 1 は密度の減少によるもので，これは電子分極にも原子分極にも関係し，温度係数は負である．第 2 は膨張とともに原子間力が減少するために，原子分極が増大することで，このような格子の独特による温度係数は正である．ルチルでは電子分極の方が原子分極よりも大きいので，密度効果は格子独特効果よりも大きく，したがって温度係数が負になるものと考えられる．

つぎに Eucken–Büchner の考えを述べる．Born[13] によればイオン結晶の誘電率はつぎの式で与えられる（§ 6 参照）．

$$\varepsilon = \varepsilon_0 + (Ze)^2 N_A \varrho / \pi f^2 M_1 M_2 \quad (88.3)$$

ε_0 は赤外波長に外挿した屈折率の 2 乗，Ze はイオンの有効電荷，N_A は Avogadro 数，ϱ は密度，f は格子の赤外線固有振動数，M_1，M_2 はイオンの原子量である．q は二原子化合物（たとえば NaCl）に対しては 1，三原子化合物（たとえば CaF_2）に対しては 2 である．

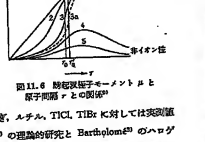

図 11.6　誘電双極子モーメント μ と原子間距離 r との関係[13]

さて上式にしたがって誘電率を計算すると，アルカリハライドに対しては実測値より小さすぎ，ルチル，TlCl，TlBr に対しては実測値よりはるかに小さな値が得られる．Kirkwood[14] の理論的研究と Bartholomé[15] のハロゲン化水素に関する実験的研究によれば，イオンの有効電荷 Z_e はイオン間距離 r に関係する．跨線[13] イオンの形成する双極子モーメント $\mu = Z_e \cdot r$ と r との関係を Eucken–Büchner は図 11.6 のように考えた．

直線 1 は点電荷とした純イオン結晶に対する仮想的なもので，曲線 2 はアルカリハライドを表わし，曲線 4，5 は等極結合性の強い物質に対するものである．曲線 3a はイオン結合と非イオン結合との境界をなし，ルチルの曲線は 3 であると考える．Eucken–Büchner は Born 式の Z_e を

$$\frac{d\mu}{dr} = Z_e + r\frac{d(Z_e)}{dr} \quad (88.4)$$

で置きかえることにより，誘電率の計算値が大きくなることを指摘した．この考えによれば，ルチルの高誘電率は曲線 3 の変曲点が平均距離 r_0 の近くにあることで説明され，また負の温度係数は曲線 3 の勾配が r_0 から r_0' になるとき減少することで説明される．

さて Born の式は誘電率の電子部分と原子部分との加算性を仮定するもので，分極を加算的とする Clausius–Mosotti の式とは大いに異なる．ルチルに対してそのいずれが正しいかは明らかでないが，もし Clausius–Mosotti 式を正しいとするならば，ルチルの高誘電率は $p = 1$ の結果とみるべきである．すなわちルチルといっても分極に関しては他の結晶と容しい挙動は示さないのである．

§89　混合物の誘電率

ルチルの粉末と松みのセラミックスまたは有機の樹脂や液体との混合物に対しては対数関係式

$$\log \varepsilon = X_1 \log \varepsilon_1 + X_2 \log \varepsilon_2 \quad (89.1)$$

が比較的成立している．ここに ε_1，ε_2 は成分物質の誘電率，X_1，X_2 はそれらの体積分率である．

一般に混合物の誘電率を成分の誘電率から計算し，また比混合物の誘電率と一成分のそれとを知って，他成分の誘電率を求めることができれば大変便利である．誘電率に限らず，種々の物理量についての種の問題は古くから多くの物理学者の研究対象となってきたが，まだ充分解決されるには至っていない．誘電率の理論は導電率と形式的に密接な関係にあるものであるが，これに関する 1926 年までの研究結果は Lichtenecker[16] や Lowry[17] により総合的にまとめられており，その後 Bruggeman[18],[19] の理論や Wachholtz–Franceson[20] の実験的研究がある．以下本節において，これらを簡単に紹介しよう．

固体誘電体論

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